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A STUDY OF THE
ADHESION AND COHESION
OF METALS

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ABSTRACT

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Research on the solid state adhesion and cohesion of metals has been conducted at National Research Corporation for the past several years. Since adhesion has been observed for metals and alloys under widely varying conditions, there is a need to distinguish among the effects of the more important variables controlling the bonding mechanism. Past research programs have found that adhesion of both metals and alloys increase with temperature, contact stress and sur-face cleanliness. This report in particular deals with the adhesion dependency on solid solubility.

author

1.0 INTRODUCTION

Research on the tendency of clean metals to adhere in ultra-high vacuum has been carried on at National Research Corporation for the past five years under NASA contracts NASr-48, NASw-734 and NASw-734, Amendment #1 and under this contract, NASw-1168.

The general objective has been to obtain additional information as to the conditions under which metals and alloys of engineering importance for space applications will adhere to one another with sufficient tenacity to hinder the relative motion or subsequent separation of components of mechanical and electrical devices used in space exploration. Such devices include bearings, solenoids, valves, slip rings, mating flanges, conical rendezvous mating surfaces, etc.

Even a small amount of adhesion would be disastrous in many cases on a space vehicle. Power on such craft is ordinarily very limited and mechanical components must operate freely. Only a few ounces of adhesion force may cause failure. There is, therefore, an important requirement for quantitative data on adhesion.

Techniques were developed in previous programs for evaluating the adhesion of metals at various temperatures by repeatedly fracturing and rejoining notched tensile specimens in ultrahigh vacuum. Two types of apparatus were used: 1) a differential expansion device, and 2) a screw drive device. The latter was found to be the most useful. The maximum adhesive force at room temperature was about 65% of the applied load for copper and 19% for 1018 steel.

No adhesion was found for hardened 52100 steel. Time in contact appears to be an important factor for copper at 200°C and above. Both 1018 steel and 52100 steel were "self-cleaning" at 500°C, the former showing repeated readings near 100% cohesion, and the latter increasing in per cent cohesion with each successive break. Except for steel at 500°C, and copper at 350°C and 400°C, adhesion dropped on each successive test.[1]

The apparatus was then modified to permit specimen positioning and the testing of up to eight specimen pairs with a single pump-down of the vacuum system. Arrangements were made for wire brushing in vacuum and/or ion bombardment just before joining. Both flat-faced and chisel-edged specimens were used, the rectangular faces or chisel edges being crossed. Tests were made at room temperature and at pressures between 10^{-8} and 10^{-9} torr. Adhesion was observed only between flat-faced, soft copper specimens when wire brushed in vacuum. The cohesion force varied from 8 to 120 lbs. after a compressive force of 2000 lbs., and appeared to depend primarily on the thoroughness of wire brushing.[2]

Further work was accomplished under NASw-734, Amendment No. 1 using the modified eight specimen test apparatus. Tests were performed on similar samples as well as dissimilar sample pairs. From the results it was concluded that adhesion of like and unlike pairs increased with temperature, applied load, surface cleaning and flow stress. Under the conditions of 10^{-8} to 10^{-9} torr vacuum and a compressive force of 2000 lbs., adhesion forces from 40 to 800 lbs. were measured.[3]

Under this contract, NASw-1168, National Research Corporation is continuing the investigation of the adhesion bonding phenomena to provide further knowledge of the bonding mechanism. The present program is divided into three phases as listed below.

PHASE I

Phase I is concerned with the characterization of the effect of an adsorbed surface film on the adhesive process. Contaminant films such as oxides will be removed by ion bombardment and the degree of desorption measured by determinations of the surface electron work function. Attempts will be made to reduce adsorbed layers to less than a monolayer to all the study of adhesion forces between truly clean surfaces.

PHASE II

Contact bonding is extremely sensitive to surface typography as affected by the smoothness and by the plasticity of surface asperities under load. Measurements of adhesion force will be correlated with plasticity for different values of hardness, asperity height and radius.

PHASE III

Mutual solid solubility of the adhering materials may also play an important role in adhesion. Both soluble and unsoluble metal pairs shall be evaluated for adhesion characteristics under conditions

in which contaminant films are absent and the compressive yield stress is exceeded.

2.0 APPROACH

It was decided to proceed initially with Phase III of the experimental program since the solubility studies would require very little modification to the existing test fixture.

The metal samples tested in this phase of the experimental program are listed in Table I.

These metal samples were tested under 10^{-8} - 10^{-9} torr vacuum and using motorized wire brushing techniques to remove the surface oxide films.

3.0 APPARATUS

The apparatus consists of a stainless steel vacuum chamber with the accessories necessary to join and separate small metal specimens in ultrahigh vacuum and to measure the forces involved. The major components were developed and used in previous programs.

Figure 1 shows the loading and force measuring devices which communicates with the inside apparatus through a flexible metal bellows. Beneath the dome in Figure 1 hangs the apparatus shown in Figures 2 and 3. Sixteen specimens (eight pairs) can be mounted on the wheels shown in Figures 2 and 3, and positioned to bring different material combinations together or to expose a given surface for cleaning.

TABLE I

METAL TEST SAMPLES

Soluble Pairs	Insoluble Pairs
Cu-Au	Cu-Ta
Cu-Ni	Ag-Fe
Ag-Au	Ag-Ni
Nb-Ta	Au-Pb
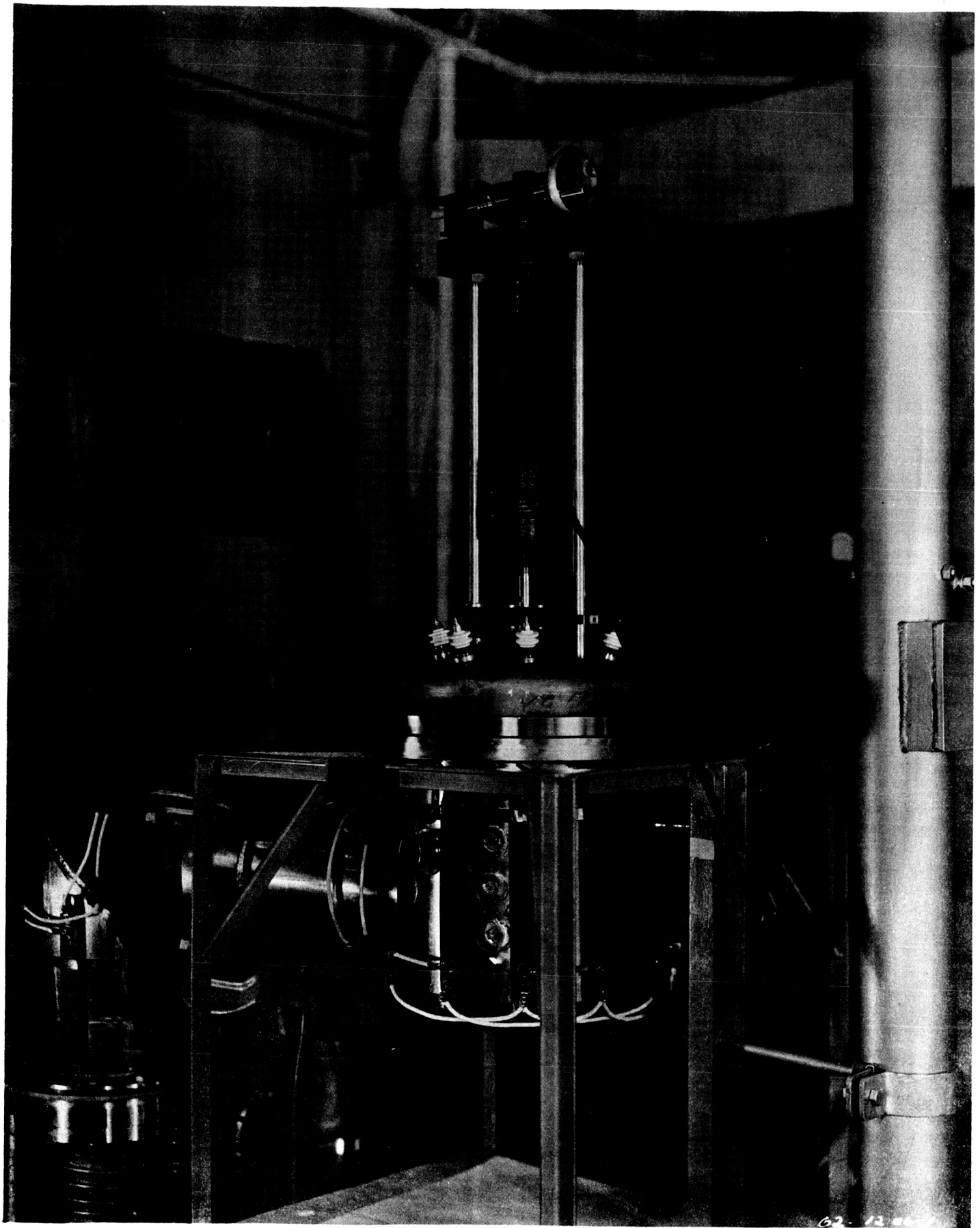
Figure 1 - Screw Drive Cohesion Testing Apparatus (outside)

WORM GEAR

BALL SCREW
DRIVE

SHEAR
PIN

LOAD CELL

SPECIMEN

BELLOWS

VACUUM
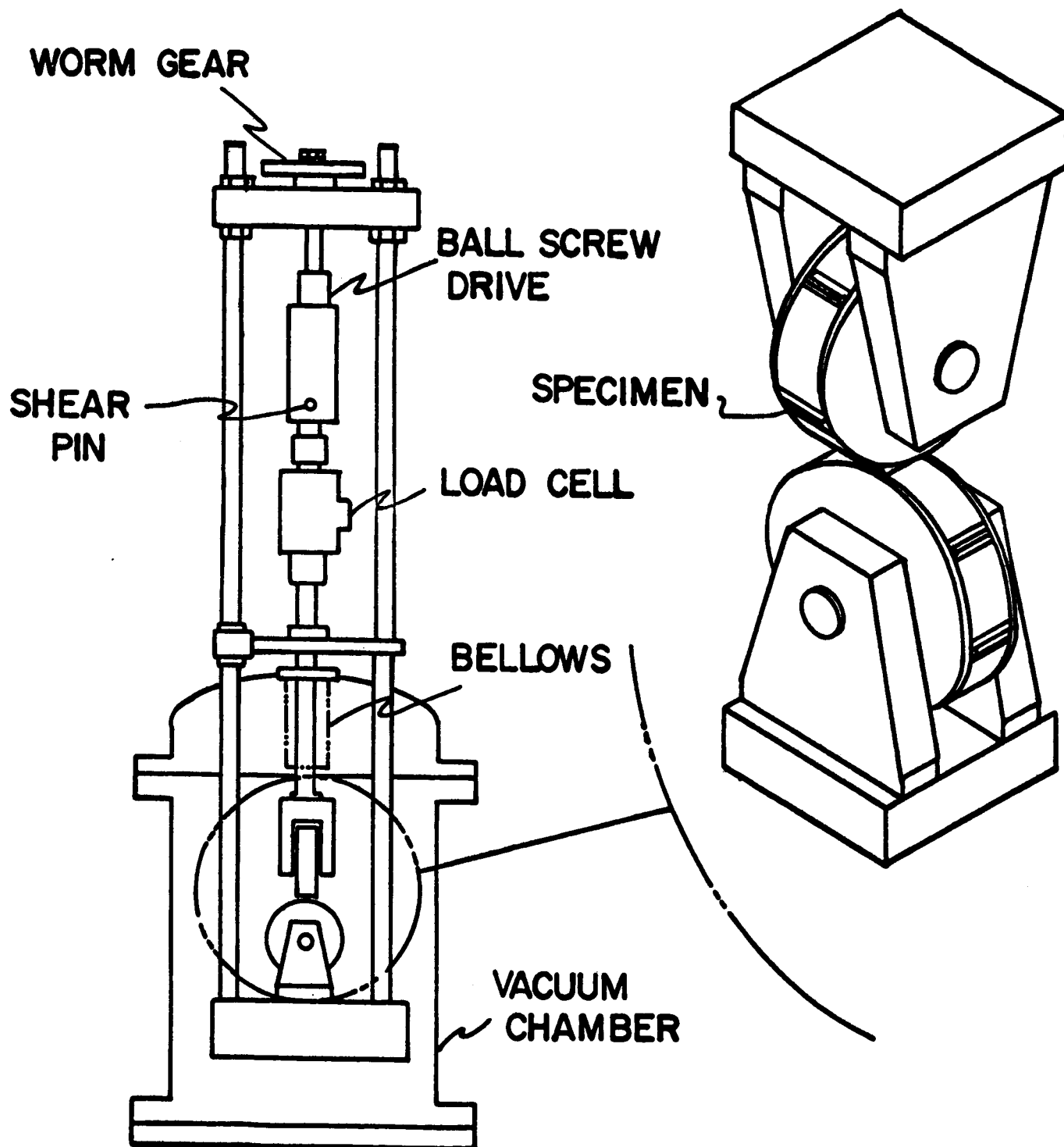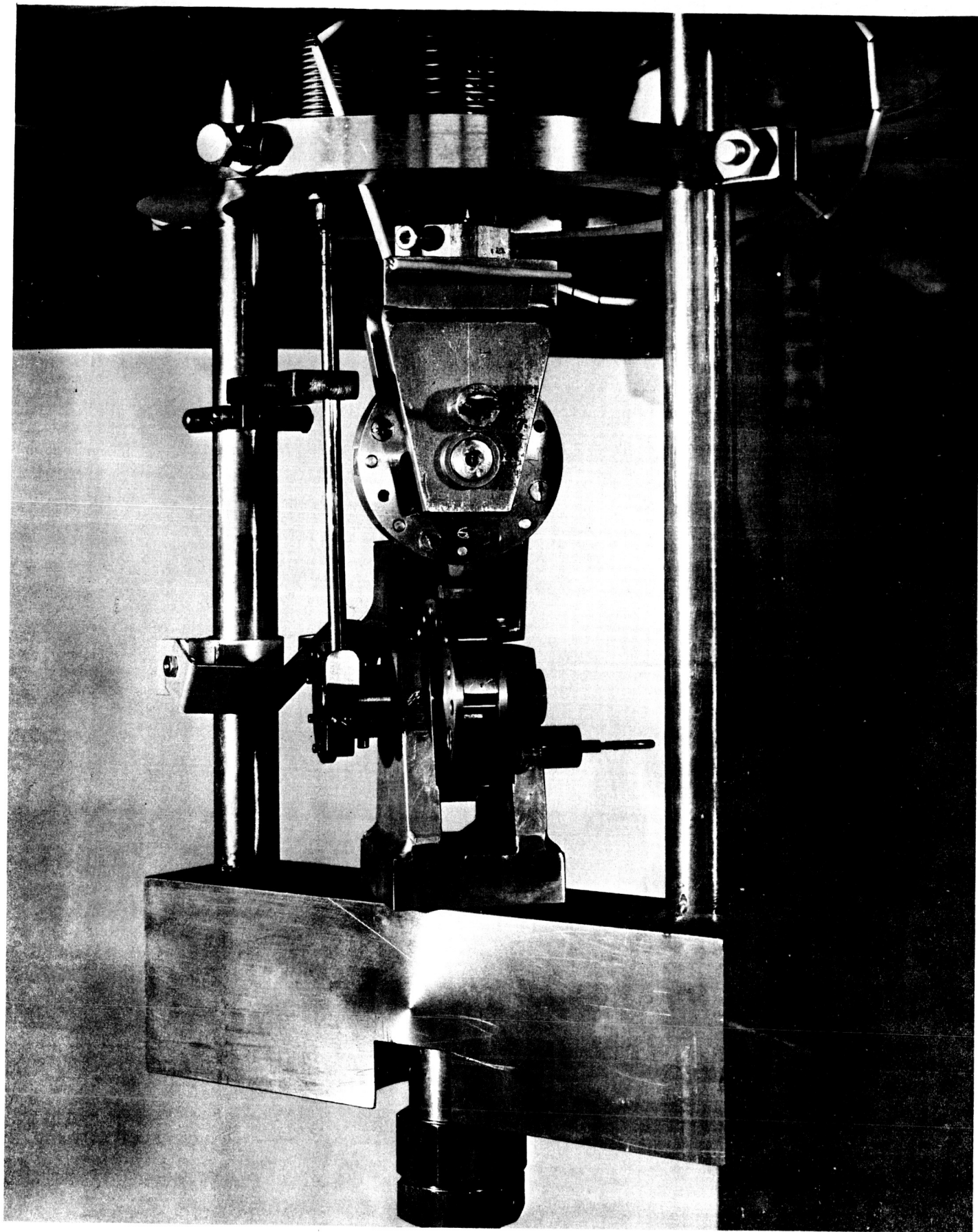CHAMBER

FIGURE 2 - COHESION TEST APPARATUS

FIGURE 3   SAMPLE HOLDER

A twelve-inch long spool piece is now located between the dome and the bowl of Figure 1 (but not shown in the figure).

The spool piece has windows for observing the specimens, and a bellows manipulator for positioning a motor-driven wire brush between specimens to clean the mating surfaces just before joining.

The entire assembly is mounted on a standard National Research Corporation ultrahigh vacuum pumping system with a 10-inch diffusion pump (HS10-4200) and a standard National Research Corporation Chevron-type liquid nitrogen trap. Concentric "O" rings cooled by a circulating refrigerant are used at the joints between the large flanges.

4.0 TEST SPECIMENS

Sixty-four test samples were fabricated to conform with the test pairs listed in Table I. The materials used were of the purest commercially available and were completely annealed. The samples were machined from 7/8" diameter bar stock which was fully annealed. However, the lead, silver, and gold materials were purchased in the dead soft condition in economical shapes to minimize machining and expense. All samples were tested for hardness and re-annealed if required before using in the adhesion experiments. The sample materials along with the pertinent data is tabulated in Table II.

The sample configuration as in previous tests was to be chisel pointed as shown in Figure 4. However, since the selected materials with the exception of tantalum and niobium have low yield stresses,

TABLE II

SPECIMEN DATA SHEET

Code	Material	Number Required	Purity	Melting Point (°C)	Condition	Hardness (BHN)	Yield Strength (psi)	Modulus of Elasticity (psi)
A	Copper (OFHC)	12	99 + Cu	1083	Full annealed 500°C 1/2 hr.	45	9000	16×10^6
B	Silver	12	99.9+Ag	960	Dead soft	40	7900	16×10^6
C	Nickel	8	99 + Ni	1455	Full annealed 1050°C 1/2 hr.	110	15000	30×10^6
D	Tantalum	8	99.9+Ta	3000	Full annealed 1050°C 1/2 hr.	79	39000	27×10^6
E	Niobium	4	99.7+Nb	2400	Full annealed 1050°C 1/2 hr.	35	32000	15×10^6
F	Iron	4	99.8+Fe	1540	Full annealed 700°C 1/2 hr.	75	18000	28×10^6
G	Lead	4	99.9+Pb	327	Dead soft	15	1200	2.6×10^6
H	Gold	12	99.9+Au	1063	Dead soft	25	2000	12×10^6

BLOCK TYPE

CHISEL TYPE

$\frac{1}{2}''$

$\frac{3}{8}''$
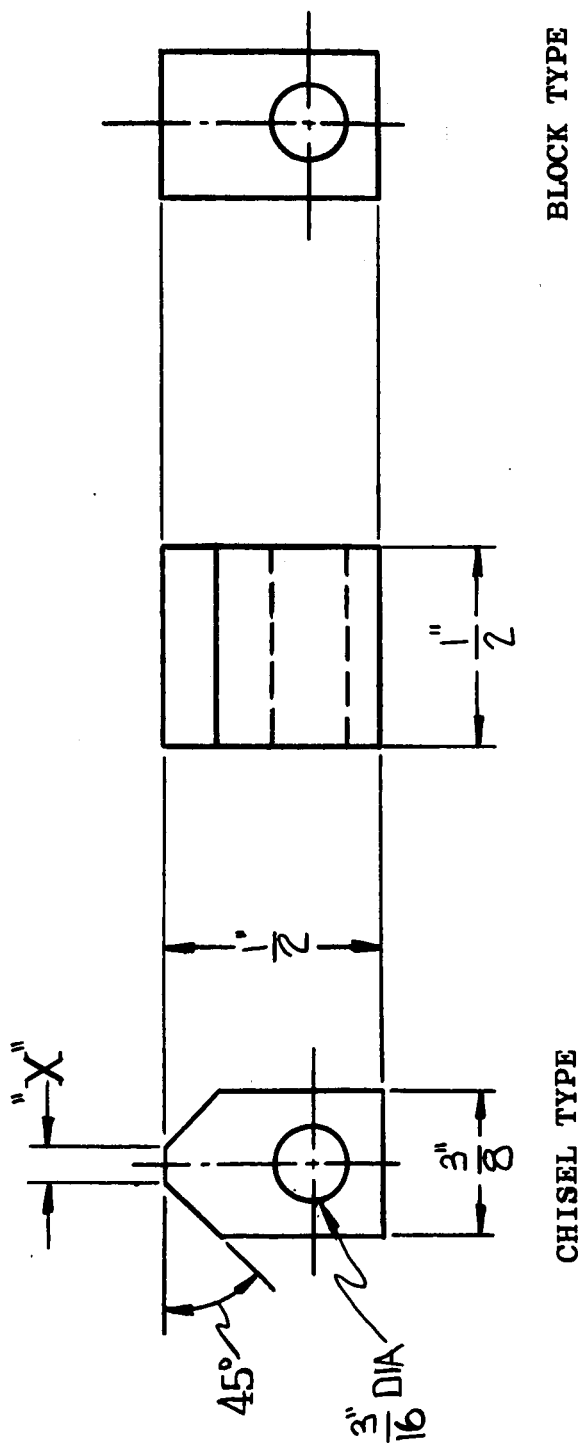
"X"

45°

$\frac{3}{16}''$ DIA

FIGURE 4

ADHESION SPECIMENS

it was decided to use a block type specimen, thus taking advantage
of the maximum surface area. The block type specimen was used
when the "X" dimension in Figure 4 approaches 3/8 inch.

5.0 EXPERIMENTAL PROCEDURE

5.1. PreVacuum Operation

The material specimens were fabricated and treated as pre-
viously specified. The samples were then tested for hardness and
if required heat treated to yield the fully annealed condition.
The face widths were measured and adjusted to produce a contact
area that would allow the load cell to operate about midrange. The
contacting surfaces were then prepared by mounting three specimens
at a time in a special jig and polishing with successively finer
grinding alundum or emery paper. The three specimen faces formed a
flat plane in the polishing jig and the abrasive paper was supported
on a lapping table. This insured specimen flatness. Just prior to
mounting in the test wheel, each specimen was given one dry run on
No. 000 paper followed by a ethyl alcohol cleaning and air dry process.
The samples were then mounted in the test wheel and the vacuum
system closed.

5.2. Test Operation

During a typical operation the system was baked out at 200°C
under vacuum for times ranging up to 48 hours, using a number of
tungsten-quartz lamps inside the vacuum vessel as a heat source. When

the pressure had stabilized at about 10^{-8} torr, a refrigeration system was turned on to cool the vessel walls and gaskets. The pressure then fell to the 10^{-9} range, typically 4 to 6 x 10^{-9} at the end of 24 hours.

To maintain the temperature of the specimens, the heat input to the lamps was controlled and the temperature was measured by thermocouples. The individual specimens were wire brushed for measured times, typically between 30 seconds and 5 minutes. During brushing of the specimens, pressure bursts occurred which yielded pressure transients up to 2 x 10^{-8} torr. This effect was more observable on the copper specimens. The specimens were then positioned to face each other and the pair to be tested were forced into contact and loaded to the desired stress level. (Relaxation and creep later reduced this load, as much as 10% in some cases.) The time between cleaning and contact was measured and found to be reasonably constant at about 14 to 19 seconds at 4 to 6 x 10^{-9} torr. This product, 2 x 10^{-9} torr-minutes, is below the 1/10th monolayer formation time and thus assures contact between clean metal surfaces. The load was left on for 5 minutes for each pair and the measured breakaway load was then recorded on pulling the specimens apart using a Sanborn recorder and SR-4 strain gauges in a Baldwin load cell as shown in Figure 2.

5.3. Post Vacuum Operation

Upon completion of the test the vacuum system was released with argon to an atmosphere and the test samples carefully removed and

packaged. Each specimen pair was optically examined to determine the effects of the bonding.

6.0 EXPERIMENTAL PROGRAM

6.1. Preliminary Adhesion Test

The first tests performed were accomplished using fully annealed copper sample pairs. These tests were performed to check out the equipment and techniques and to familiarize the personnel with the equipment. Copper samples were chosen since the most complete adhesion data has been obtained for copper. These tests were designed to re-evaluate past tests and were set up to verify that the adhesion force is a function of applied stress and surface cleaning.

The first series (six sample pairs) was designed to determine the adhesion coefficient as a function of applied load. Stress levels from 50 to 250 per cent of the yield stress were used. The fixed parameters were; 6×10^{-9} torr vacuum level, two minutes cleaning time and five minutes loading time at a temperature of 25°C. The results were that the adhesion coefficient defined as the ratio of bonding force to applied load increased from 0.05 for 100% yield stress loading to 0.20 for 250% yield stress loading as shown in Table III and Figure 5.

The second series (four sample pairs) was designed to determine the adhesion coefficient as a function of cleaning time. Brushing times of 0.5, 2 and 5 minutes were selected as variables. The fixed

TABLE III

PRELIMINARY COHESION TEST SERIES

Test No.	Test Mat'l. Combination	Yield Strength (psi)	$C=\dfrac{\text{applied stress}}{\text{yield stress}}$	Contact Area (inch²)	Pressure (torr)	Temp. (°C)	Cleaning Time (min)	Contact Time (min)	Loading Time (sec)	Applied Load (lbs)	Relaxation Load (lbs)	Break Away Load (lbs)	$\alpha=\dfrac{\text{Break Away Load}}{\text{Applied Load}}$
1	Cu-Cu	10,000	0.50	0.078	6×10^{-9}	25	2	17	5	390	10	0	0.00
2	Cu-Cu	10,000	1.00	0.085	6×10^{-9}	25	2	16	5	850	20	40	0.05
3	Cu-Cu	10,000	1.25	0.078	6×10^{-9}	25	2	17	5	940	40	65	0.07
4	Cu-Cu	10,000	1.50	0.087	6×10^{-9}	25	2	15	5	1300	40	110	0.09
5	Cu-Cu	10,000	2.00	0.076	6×10^{-9}	25	2	16	5	1700	60	260	0.15
6	Cu-Cu	10,000	2.50	0.081	6×10^{-9}	25	2	17	5	2500	90	430	0.20
7	Cu-Cu	10,000	1.25	0.078	6×10^{-9}	25	5	17	5	940	40	200	0.21
8	Cu-Cu	10,000	1.50	0.087	6×10^{-9}	25	5	16	5	1300	50	400	0.31
9	Cu-Cu	10,000	1.50	0.087	6×10^{-9}	25	0.5	16	5	1300	50	30	0.02
10	Cu-Cu	10,000	1.25	0.78	6×10^{-9}	25	0.5	17	5	990	40	10	0.01

FIGURE   5

$\alpha = \dfrac{\text{ADHESION FORCE}}{\text{APPLIED FORCE}}$

$C= \dfrac{\text{APPLIED STRESS}}{\text{YIELD   STRESS}}$
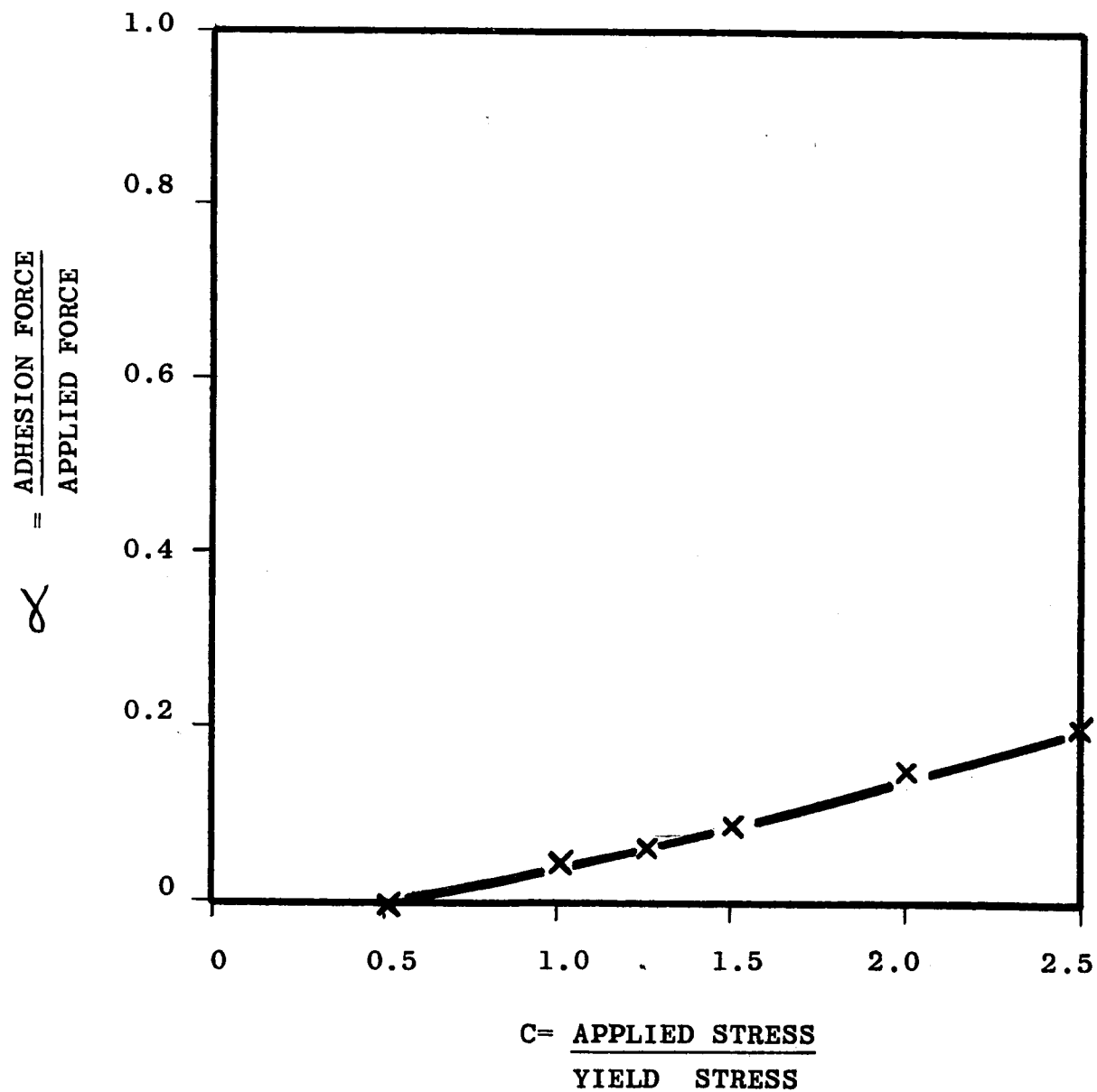
ADHESION OF COPPER SPECIMENS WITH

INCREASE LOADING (DATA FROM TABLE III)

parameters were; 6×10^{-9} torr vacuum, stress levels at 125% and 150% of yield stress, 5 minutes holding time and temperature at 25°C. The results were that the adhesion coefficient increased from 0.01 to 0.21 for 125% yield stress loading and 0.02 to 0.31 for 150% yield stress loading as shown in Table III and Figure 6.

6.2. Adhesion - Mutual Solubility Test

Two types of metals pairs were tested in the solubility test program; metals with extremely limited mutual solubility and metals which form extensive solid solutions at room temperature. Metals of similar FCC and BCC crystal structure were selected to minimize lattice misorientation and strain accommodation effects. The materials tested are listed in Table I.

6.2.1. Solubility Test

The materials listed as soluble in Table I are known to form complete solid solutions at room temperature. The tests were designed to determine the adhesion coefficient as a function of solubility and lattice parameters. Parameters held constant were; vacuum level at 5×10^{-9} to 1×10^{-8} torr range, cleaning time at 2 minutes and loading time at 5 minutes. Other variables such as stress level and temperature were varied to determine if dissimilar metal pairs followed the adhesion trends of similar pairs. Adhesion was obtainable on all tested samples except the tantalum-niobium pair. These metals form tenacious oxides and require more extensive cleaning techniques to produce oxide-free surfaces. Data on the

FIGURE 6
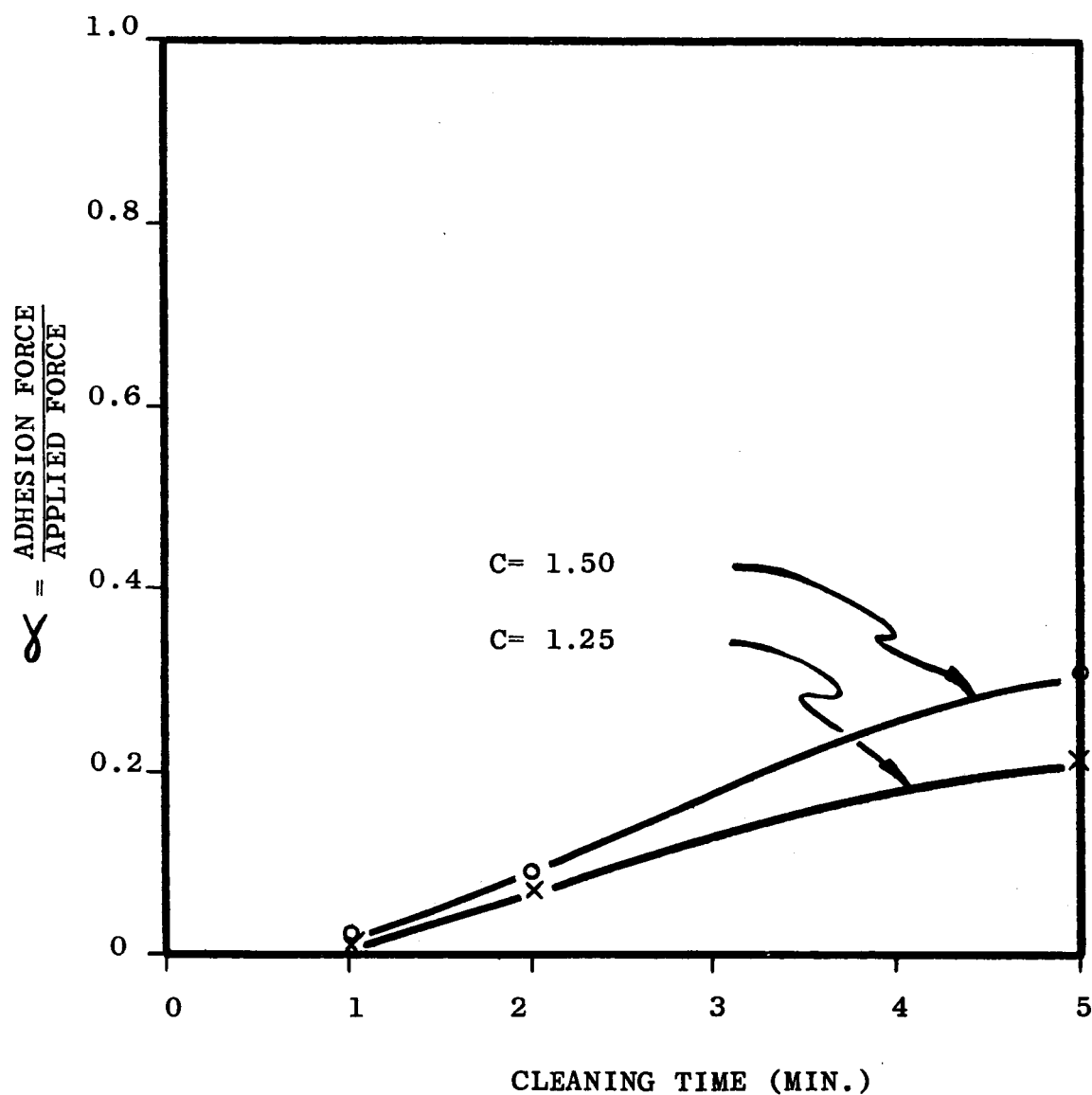
ADHESION AS A FUNCTION OF CLEANING FOR
COPPER SPECIMENS USING WIRE BRUSHING
TECHNIQUES (DATA FROM TABLE III)

soluble test pairs is tabulated in Table IV.

6.2.2. Insolubility Test

The insoluble metal pairs listed in Table I are all known to have less than 0.1% solubility in each other. Tests on these materials were designed to be used as a comparison for the solid solution metal pairs. The fixed and varied parameters were the same as those used in the solubility tests. However, it was expected that very little adhesion would occur with these pairs. Adhesion coefficients were obtained for all pairs with maximum coefficient of 0.6 for the lead-gold combination. Data on the insoluble adhesion test pairs is tabulated in Table V.

7.0 DISCUSSION OF RESULTS

The comparative data taken on soluble and insoluble metals showed no correlation between adhesion and lattice solubility. However, it does appear that adhesion is related to plastic loading, temperature and yield strength as shown in Figure 7.

The relative insensitivity of adhesion bonding at 25 to 150°C to bulk lattice solubility levels may be attributed to the extreme importance of local atomic arrangements on the surface layers in determining the density of bonds across the interface. Increases in the effective lattice parameter and local disorder at the surface may be expected due to the anistropy of atomic packing and bond distribution. In this way, solute atoms of greatly differing size

TABLE IV

SOLUBLE ADHESION TEST SERIES

Test No.	Test Mat'l. Combination	Yield Strength (psi)	$C=\dfrac{\text{applied stress}}{\text{yield stress}}$	Contact Area (inch^2)	Pressure (torr)	Temp. (°C)	Cleaning Time (min)	Contact Time (min)	Loading Time (sec)	Applied Load (lbs)	Relaxation Load (lbs)	Break Away Load (lbs)	Break Away Load $=\dfrac{\text{Away}}{\text{Applied Load}}$
1*	Cu-Ni	9000	1.25	0.131	6×10^{-9}	25	2	15	5	1470	20	0	0.00
2	Cu-Ni	9000	1.25	0.131	5×10^{-9}	25	2	17	5	1470	10	180	0.12
3	Cu-Ni	9000	1.50	0.131	6×10^{-9}	25	2	15	5	1770	50	400	0.23
4	Cu-Ni	9000	1.50	0.131	9×10^{-9}	150	2	19	5	1770	50	740	0.42
5	Au-Cu	2000	1.25	0.148	6×10^{-9}	25	2	15	5	370	10	24	0.07
6	Au-Cu	2000	1.50	0.141	6×10^{-9}	25	2	14	5	425	10	90	0.21
7	Au-Cu	2000	1.50	0.145	9×10^{-9}	150	2	18	5	435	25	210	0.48
8	Au-Ag	2000	1.25	0.145	6×10^{-9}	25	2	15	5	360	20	38	0.11
9	Au-Ag	2000	1.50	0.147	7×10^{-9}	25	2	15	5	440	30	10	0.02
10	Au-Ag	2000	1.50	0.145	7×10^{-9}	25	2	18	5	440	40	40	0.09
11	Au-Ag	2000	2.00	0.147	7×10^{-9}	25	2	15	5	580	40	39	0.07
12	Au-Ag	2000	1.50	0.143	1×10^{-8}	150	2	16	5	430	10	180	0.42
13	Nb-Ta	32000	0.85	0.070	6×10^{-9}	25	2	18	5	1925	60	0	0.00
14	Nb-Ta	32000	1.00	0.070	5×10^{-9}	25	2	15	5	2300	60	0	0.00
15	Nb-Ta	32000	1.50	0.040	4×10^{-9}	25	2	19	5	1920	80	0	0.00
16	Nb-Ta	32000	2.00	0.031	4×10^{-9}	25	2	15	5	1980	100	5	0.01
17	Nb-Ta	32000	1.00	0.070	1×10^{-8}	150	2	15	5	2300	80	0	0.00
18	Nb-Ta	32000	1.50	0.038	1×10^{-8}	150	2	15	5	1820	120	40	0.02

* Inadequate brushing.

TABLE V

INSOLUBLE ADHESION TEST SERIES

Test No.	Test Mat'l. Combination	Yield Strength (psi)	$C = \dfrac{\text{applied stress}}{\text{yield stress}}$	Contact Area (inch^2)	Pressure (torr)	Temp. (°C)	Cleaning Time (min)	Contact Time (min)	Loading Time (sec)	Applied Load (lbs)	Relaxation Load (lbs)	Break Away Load (lbs)	$\dfrac{\text{Break Away Load}}{\text{Applied Load}}$ (%)
19	Cu-Ta	9000	1.25	0.131	6×10^{-9}	25	2	16	5	1470	40	70	0.05
20	Cu-Ta	9000	1.50	0.131	6×10^{-9}	25	2	14	5	1770	20	20	0.01
21	Cu-Ta	9000	1.50	0.131	7×10^{-9}	25	2	16	5	1770	30	130	0.07
22	Cu-Ta	9000	1.50	0.131	1×10^{-8}	150	2	14	5	1770	50	600	0.34
23	Ag-Fe	7900	1.25	0.131	7×10^{-9}	25	2	16	5	1310	30	140	0.11
24	Ag-Fe	7900	1.50	0.131	7×10^{-9}	25	2	14	5	1550	50	280	0.18
25	Ag-Fe	7900	1.50	0.131	1×10^{-8}	150	2	16	5	1550	60	380	0.25
26	Ag-Ni	7900	1.25	0.131	6×10^{-9}	25	2	17	5	1310	30	200	0.15
27*	Ag-Ni	7900	1.50	0.131	7×10^{-9}	25	2	60	5	1550	40	40	0.03
28	Ag-Ni	7900	1.50	0.131	4×10^{-9}	25	2	15	5	1550	50	260	0.17
29	Ag-Ni	7900	1.50	0.131	1×10^{-8}	150	2	16	5	1550	40	260	0.17
30	Pb-Au	1200	1.25	0.141	1×10^{-8}	25	2	17	5	210	30	120	0.57
31	Pb-Au	1200	1.50	0.142	1×10^{-8}	25	2	18	5	253	35	150	0.59
32**	Pb-Au	1200	1.50	0.141	5×10^{-8}	115	2	18	5	255	45	110	0.43

* Brush broke.

** Lead sample elongated did not break at cold weld joint.

FIGURE 7

SAMPLE PAIR MATERIALS
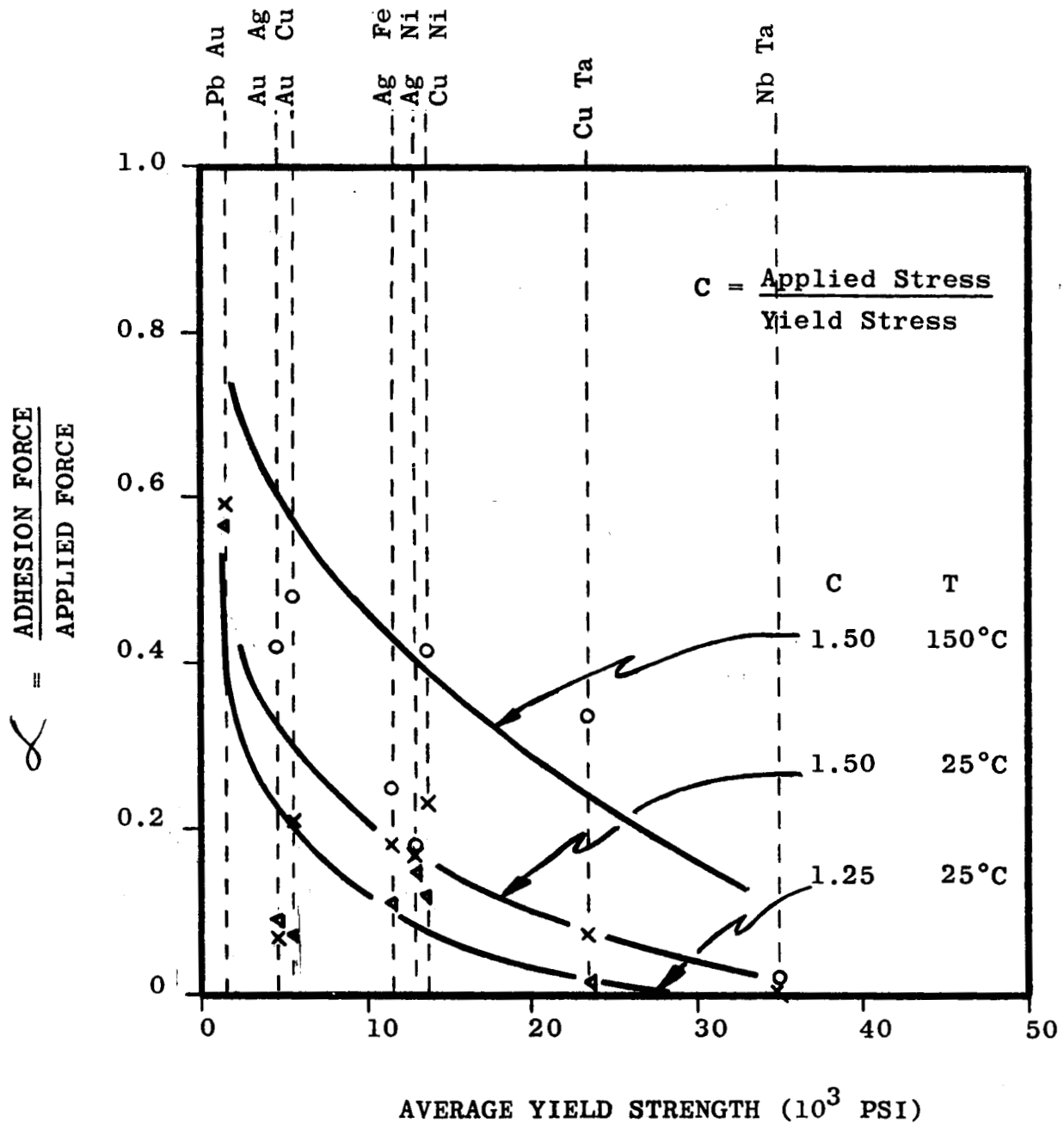
ADHESION COEFFICIENT AS A FUNCTION IN
YIELD STRESS FOR VARIOUS SAMPLE MATERIALS
(DATA FROM TABLES IV & V)

and electrochemical activity may be accepted substitutionally or interstitially in the solvent lattice within a few atomic layers of the surface, but would be rejected in the bulk lattice. Thermal diffusion across the bonded interface of chemically insoluble metals would lead to intermetallic compound or terminal phase precipitation and segregation within the lattice.

In all cases with both soluble and insoluble sample pairs, the adhesion force increased with load and temperature. The exception was the Ta-Nb and Au-Ag combinations. The Ta-Nb combination showed no adhesion coefficient at 25°C with varying load. This can be accounted for by the tenacious oxides formed on both Ta and Nb metals. These oxides are difficult to remove and require better cleaning techniques. The Ag-Au combination showed some adhesion force at 25°C but it remained relatively constant with increased loading.

Another parameter which appeared to influence the ability to adhere is the flow stress or hardness of the materials. The data as in past reports indicated that adhesion increases with decreasing hardness or flow stress level.

To summarize, adhesion tends to increase with temperature, surface cleanliness, stress level, and to decrease with hardness. However, no correlation was observed between adhesion and mutual solubility or lattice parameter.

8.0 FUTURE PLANS

Plans have been made to start Phase I of the test program. This requires the use of two ion-guns for cleaning of the sample faces. The system is presently being modified to accommodate the two guns and changes are being made in the sample positioning device.

The test series for Phase I is tentatively scheduled for August at which time the system should be completely modified and checked out.

REFERENCES

1. National Research Corp., Cambridge, Mass., "INVESTIGATION
    OF ADHESION AND COHESION OF METALS IN ULTRAHIGH VACUUM,
    First Annual Summary Report, June 15, 1961 to July 31, 1962".
    John L. Ham, September 7, 1962, (N62-17772), (NASA Contract
    NASr-48; NRC Project No. 41-1-0121).

2. National Research Corp., Cambridge, Mass., " INVESTIGATION
    OF ADHESION AND COHESION OF METALS IN ULTRAHIGH VACUUM,
    Final Report, March 1, 1963 to November 1, 1963". John L. Ham,
    November 27, 1963, (NASA Contract NASw-734; NRC Project No.
    81-1-0101).

3. National Research Corporation, Cambridge, Mass., "ADHESION
    AND COHESION OF METALS, Final Technical Report, March 24, 1964
    to January 1, 1965". L.R.Allen, P.L.Vitkus,J.L.Ham, and
    F.J.Brock, April 1, 1965, (NASA Contract NASw-734, Amendment
    No. 1, NRC Project No. 82-1-0219).